IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant(s): Donald R. Huffman, et al.

Examiner: Hendrickson

Serial No.: 07/580,246

Art Unit: 1754

Filed: September, 10th 1990

Docket: 7913Z

For: NEW FORM OF CARBON

Assistant Commissioner for Patents
Washington, DC 20231

DECLARATION OF Raouf O. Loutfy

I, Raouf Loutfy, hereby declare:

1. I reside at 6507 N. Ventana Canyon Drive, Tucson, Arizona, USA.

2. I earned a B.Sc. degree in 1964 in Applied Chemistry from Cairo University, a M. Sc. in 1966 in Solid State Sciences from the American University, a Ph.D in 1971 in Physical Chemistry from University of Western Ontario, and a Diploma in Business Administration in 1976 from McGill University.

3. Between 1977 and 1981 I was employed as a group leader at Argonne National Laboratory in the Chemical Engineering Division.

4. Between 1981 and 1985 I was employed as Research Advisor of ARCO Metals/ARCO Chemical developing advanced material technologies.

5. Between 1985 to present I have been employed as president of MER Corporation, and since 1990 I have been involved in the commercial scale-up of fullerenes, and in developing applications for fullerenes.

6. I have published many articles, two books, and contributed a chapter in Encyclopedia of Technology on the fullerene production, and on applications of fullerenes. I have been an invited speaker to many technical and investment conferences as an expert in the technology and commercialization of fullerenes. I have received the prestigious Tibbetts award in 2001 from the Small Business Administration (SBA) for the commercialization of fullerenes.

For the convenience of the patent and trademark office, I have attached hereto as Exhibit 1 my curriculum vitae, which describes my credentials and demonstrates my expertise in the area of fullerenes.

7. I am intimately familiar with the published literature concerning fullerenes and I am personally involved in the research and development of new methods of production and applications for fullerenes including C_{60}, C_{70} and nanotubes.

8. I am informed by Mark J. Cohen, Esq., the attorney handling the prosecution of the subject United States patent application that a question has arisen concerning the use

of the term "macroscopic amounts" as applied to the fullerene products, e.g. C_{60} and C_{70} that are produced by the methods invented by Drs. Huffman and Kratschmer as described in the underlying application.

9. As set forth above, I have been professionally involved in the field of fullerenes development and manufacture since Dr. Huffman and Kratschmer disclosed their novel process for producing macroscopic quantities, i.e., visible quantities of fullerenes, e.g., C_{60} and C_{70}, as disclosed in the subject patent application and in their publication in Nature (1990) identified elsewhere in this Declaration and I am familiar with the use of the term "macroscopic quantities" in this field and, in general, in the scientific and academic communities.

10. I have read and reviewed the subject patent application, including the pending claims. It is my understanding that the subject application discloses, among other things, a process of producing fullerenes including C_{60} and C_{70} in macroscopic amounts and that, inter alia, U.S. Serial No. 07/580,246 includes claims directed to macroscopic quantities of C_{60} and C_{70}.

11. Although the subject patent application of Dr. Kratschmer and Huffman does not expressly use the term "macroscopic amounts" to describe the amounts of fullerenes, e.g., C_{60} and C_{70} first isolated by them, in accordance with the teaching of the process described therein, the fullerenes, e.g., C_{60} and C_{70}, that were prepared in accordance with the process described herein, were produced in measurable amounts that were visible to

them, and it is my professional opinion that these amounts are inherently amounts definable by the term "macroscopic amounts".

12. Part of our research and development at MER involves the use of various arc processes, plasma processes, and high temperature process to produce fine metal, ceramic, and carbon powders. In the course of this research and development work, MER has since from about 1985 to the present maintained equipment and capabilities that could have produced soot. I am quite familiar with the principal of operations and the use of such equipment. At MER, I regularly produced soot under arc, plasma, and high temperature conditions for various applications. I am also familiar with the work of the Nobel Laureate Sir Harry Kroto et al. al. published in Nature volume 318, No. 6042, 14-20 November 1985. p162. In this publication they report the detection of remarkably stable cluster of 60 carbon "atom" using time-of-flight mass spectrometry in the vapor phase. However, they never isolated or recovered visible particle of C_{60} and C_{70}, and did not disclose a process that would teach or lead others to do so. It is noted they reported the following "If a large-scale synthetic route to this C_{60} species can be found, the chemical and practical value of the substance may prove extremely high".

13. On or about September 18, 1990, MER received and executed confidentiality agreement from RCT regarding disclosure to MER of Huffman's et al. al. discoveries.

14. On or about September 20, 1990, Dr. Huffman of U of A and Mr. Jacobs of RCT visited MER Corporation to further discuss proactive strategy concerning an arrangement

to license from RCT under the Huffman et. al. invention to produce fullerenes at MER and to capitalize on the fullerene discovery of Dr. Huffman et al., and to discuss in details the production and the separation of fullerenes from the vaporized carbon utilizing the Huffman et al. discoveries.

15. On October 9, 1990, Dr. Withers, CEO of MER, and I visited Dr. Huffman's laboratory and observed his apparatus and witnessed the operation of his apparatus for vaporization of carbon, and production and collection of soot containing fullerenes. In many respects, Dr. Huffman's apparatus was very similar to much of the equipment at MER; however, MER had not used their equipment to produce fullerenes prior to the Huffman et. al. invention. The operating conditions for such equipment and the realization of the existence of fullerenes in the soot were the missing elements, not only from MER's viewpoint, but also from that of all other researchers prior to the Huffman et. al. invention. The equipment of MER and others will not produce inherently fullerene and even if produced it was not known that it could be recovered as it was later taught by Dr. Kratschmer and Huffman. All researchers in this field either produced soot under high vacuum conditions, without sufficient concentration of carbon vapor, at low pressure, or without sufficient cooling or combination of these conditions to produce recoverable fullerenes in the soot. These conditions resulted in product that does not contain fullerenes. Even though it appears simple to the uninformed, especially in hindsight, the process of Dr. Kratschmer and Huffman as described in the subject application, is a remarkable discovery, which produced a high density of vapor of carbon as described on page 4 of the subject application which resulted in the formation of

macroscopic amounts of fullerenes by their method. From 1985, when Dr. Smally et. al. at Rice University discovered the existence of C_{60} and C_{70} atoms by spectrographic analysis of a vapor (see paragraph 12 above), until Dr. Huffman et. al. published their discovery in 1990 no one else realized how to produce and recover macroscopic quantities of these fullerenes, despite the availability of equipment that could have been used for this purpose.

16. I directed experiments to produce fullerene soot at MER using equipment at MER, and we also built additional low vacuum equipment based on the Kratschmer and Huffman method described in the subject application and produced macroscopic quantities of soot and fullerene. We separated the fullerene according to their teaching. From a similar apparatus as that described in the subject application, but operated for sufficient time, we were able to offer for sale to the research community fullerenes including pure (90%+) C_{60} and C_{70} by November 1990.

We have since developed computer control, fully automated reactor equipment using Kratschmer and Huffman's teaching in the subject application and we optimized the process to maximize the yield of fullerenes.

17. I repeated exactly the Huffman et. al. process according to the teaching described in the subject application including example 1 using ¼ inch in diameter graphite rod, at 100 torr Helium, using 100 ampere dc current. This graphite vaporized, and the vapor was condensed on a water cooled surface. The vaporization was performed for 50 minutes using about 17 cm length of the graphite rod and produced 12 gram of soot. The

fullerenes were recovered using tolerene and the amount of fullerene was determined. The yield of fullerene was about 8 to 10%. Accordingly, the total recoverable fullerenes was over 1.2 grams with over 900 mg of C_{60} and over 200 mg of C_{70} and the remaining other fullerenes.

According to the teaching in the subject application where they vaporized a ¼" graphite rod with 1 cm length, the inventor must have produced at least about 600 mg of soot that contains admixture of at least 63 mg of fullerenes that contain at least about 50 mg of C_{60} and at least about 10 mg of C_{70}. A 600-mg quantity of soot certainly can be seen by the naked eye, as also indicated by the inventor that "heavy block coating on collecting substrates and/or on the walls of the chamber which can be easily scraped off for the recovery step." Also, the 60 mg of fullerene certainly can be seen by the naked eye and it is measurable. Furthermore, the 45 and 10 mg of C_{60} and C_{70} respectively are also measurable, in today modern laboratory facility amount as low as 0.1 mg can be measured, and can be seen by the naked eye.

The same conclusion can be reached by simply calculating the mass of the rod vaporized in Kratschmer et al. subject application, including example 1, which is easily determined from the diameter of the graphite rod they used (1/4"), the length (1 cm), and typical density of the type of graphite used for graphite vaporization (2.0 g/cc). This calculation estimates that about 633 mg of soot containing fullerenes was produced by Kratschmer et al., which is certainly macroscopic and in agreement with the above-presented experimental data.

Moreover, if a longer graphite rod were used, the amount of C_{60}, C_{70} and other fullerenes produced would even be greater, as shown herein above.

18. It is in my opinion that the inventors of this subject application were the first to isolate and recover a measurable or macroscopic amount of fullurenes, and to teach others to do so. Their description in the application is clearly understood by ordinary skilled artisans, and when repeated by us allowed us to produce visible, measurable commercial quantities of fullerene product, commonly described as "macroscopic quantities".

19. It is also in my opinion that the inventors produced "macroscopic amounts" as used in the claims as clearly understood by the ordinary skilled artisan, and supported by the above. The term is commonly used to connote that amount which can be seen easily with the naked eye. This opinion is corroborated by the usage of this term by others in the fullerene field to describe the fullerene product produced by the process invented by Drs. Kratschmer and Huffman, for example, see the statement appearing at Column 1, lines 58-61 in U.S. Patent No. 6,077,401 issued June 20, 2000, on an application filed August 15, 1994, which is as follows:

> "All of these applications have been discovered since the first macroscopic amounts of the most common fullerene, C_{60}, were isolated in 1990 [Kratschmer, et al., Nature 347, 354 (1990)]."

The cited quote from the Fields et. al. patent also corroborates the common understanding of those familiar with the fullerene art that they were the first persons to produce fullerenes in macroscopic quantities and to show others how to do so.

For the convenience of the patent and trademark office, I have attached hereto as Exhibit 2 the cited patent.

I further declare that all statements made herein of my own knowledge are true, and all statements made on information and belief are believed to be true. I acknowledge that willful false statement and the like are punishable by fine or imprisonment or both (18 U.S.C. §1001) and may jeopardize the validity of the application or any patent issuing thereon.

July 16, 2002
Date

Raouf O. Loutfy, Ph.D

Signed in Tucson, Arizona